PATENT SPECIFICATION 1,013,888

1,013,888

DRAWINGS ATTACHED.

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COMPLETE SPECIFICATION.

Improvements in or relating to Methods of and Apparatus for Reacting of Fluids.

We, THE POWER-GAS CORPORATION LIMITED, a British Company, of Parkfield Works, Stockton-on-Tees, County Durham do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to methods of and apparatus for the reacting of fluids wherein fluid mixture flowing through and leaving a reaction zone is recirculated to said zone.

Recirculation of fluid mixture through a reaction zone results in mixing of said fluid mixture with the fluid reactants initially entering said reaction zone, whereby an initial reaction temperature and increased turbulence within the reaction zone are rapidly achieved, and the temperature in the reaction zone is more uniform, whether the reaction be exothermic or endothermic. Such recirculation also results in recycling through the reaction zone, whereby a high overall degree of conversion per pass is relatively low.

It is known to pass a pressurized mixture of fluid reactants through a nozzle arranged to aspire, on the ejector principle, fluid mixture leaving a reaction zone, causing mixing of such fluid mixture with the reactants and compression of the resultant mixture, thus achieving recirculation through the reaction zone.

Such an arrangement has the attraction that only a simple piece of stationary equipment, namely a nozzle arranged to operate on the ejector principle, is involved.

However, the nozzle arrangement has a major disadvantage in that it is reasonably efficient only at or near its design rating. For example, consider a nozzle arrangement designed so that, for a given rate of flow of the fluid reactants, a recirculation of the fluid mixture leaving a reaction zone is induced at the rate of twenty times the rate of flow of the reactants. Consider that the fluid reactants are passed at half the design rate of flow. The linear velocity at the throat of the nozzle will be one half of the design velocity and the fluid kinetic energy and pressure drop across the nozzle will be one quarter of the design values. The rate of recirculation of said fluid mixture will be reduced approximately proportionally to approximately five times the rate of flow of the reactants.

According to one aspect of the present invention, a method of continuously reacting fluids with one another comprises injecting a pressurized mixture of fluid reactants through at least one nozzle, having a throat with a variable flow cross-sectional area, and into a circuit, comprising a reaction zone connected by its outlet back to its inlet, to induce recirculation of the fluid mixture through said reaction zone and around said circuit, adjusting the flow cross-sectional area of said nozzle throat to obtain a desired rate of recirculation of said fluid mixture, and withdrawing a proportion of said recirculated fluid mixture as reaction product.

The flow cross-sectional area of the nozzle may be adjusted manually or automatically according to the pressure drop across the nozzle, to which the rate of recirculation is proportional.

The fluid reactants may be gaseous or liquid. Each reactant may be supplied as a stream and the separate streams are preferably mixed, at desired proportional rates of flow, near the vicinity of said nozzle. One

or more of the reactant streams may be preheated.

The reaction zone may be a free space or it may be filled with a catalyst material suitable for promoting the desired reaction.

In cases where in the reaction zone is large, for large throughputs, more than one nozzle may be provided, each having a throat with a variable flow cross-sectional area.

When the reaction is exothermic, said recirculated fluid mixture may be cooled by indirect heat exchange with a cooling medium, and when the reaction is endothermic said recirculated fluid mixture may be heated by indirect heat exchange with a heating medium.

According to another aspect of the invention, apparatus for continuously reacting fluids with one another comprises a reaction vessel which contains a reaction zone connected by its outlet back to its inlet to form a circuit, at least one fluid-reactant inflow connection member terminating in an injector nozzle arranged in said circuit for the injection of a mixture of fluid reactants into said reaction zone in such a manner as to induce the recirculation of fluid mixture around said circuit and through said reaction zone, means for varying the flow cross-sectional area at the throat of said nozzle and an outflow connection member from said circuit for reaction product.

Said means for varying the throat area of the nozzle may comprise a suitably shaped, for example conical, nozzle constricting member attached to a spindle concentric with the axis of the nozzle, said spindle being movable along said axis towards and away from the throat of the nozzle, so that the flow cross-sectional area at the throat of the nozzle may be varied from fully closed to fully open. Said spindle may be screw threaded and attached to a hand wheel for manual adjustment or it may be connected to a motor of an automatic control system for automatic adjustment.

The pressure drop across the nozzle can be measured by a suitably connected pressure gauge or differential pressure meter. The spindle of the nozzle constricting member may be adjusted either manually or automatically to maintain a predetermined pressure drop across the nozzle, corresponding to a desired rate of recirculation of said fluid mixture.

The nozzle may form a part of said inflow connection member, or it may be rigidly and detachably attached to said member, so that different nozzles of different throat areas may be installed, as required.

The reaction vessel may be suitably partitioned so that the circuit for recirculation is wholly within the reaction vessel.

In one embodiment of the apparatus, the reaction vessel has an inner tubular member concentric with the longitudinal axis of the vessel and open at both ends, said nozzle being directed concentrically towards an inlet end of said subular member for inducinlet end of said tubular member for inducsuch fluid returning to said inlet end of said tubular member through the annular space between the outer wall of the vessel and said tubular member. The reaction zone is contained principally within said tubular member but may extend to said annular space. Said inner tubular member may incorporate a convergent section at its inlet end and may incorporate a divergent section at its other open end.

In another embodiment of the apparatus, the reaction vessel may comprise a shell with tube plates rigidly attached thereto at opposite ends and a number of open-ended tubes expanded into or otherwise rigidly attached to said tube plates and extending from one tube plate to the other, a central tube serving for recirculation of fluid mixture and outer tubes constituting the reaction zone, said nozzle being inserted into said reaction vessel to be directed towards said central tube and concentric therewith, the reaction vessel having inlet and outlet connections for a cooling or heating medium for passage through the space outside said tubes and between the tube plates. Said outer tubes may contain a suitable catalyst material.

In a further embodiment of the apparatus, the whole or a major part of the reaction vessel constitutes the reaction zone and a recirculation conduit branches from said outflow connection member and leads back to an inlet to the reaction vessel, said nozzle being directed into said recirculation conduit, preferably close to the inlet to the reaction zone, for the recirculation of fluid mixture. The reaction vessel may contain a suitable catalyist material. A heat exchanger may be incorporated in said recirculation conduit, whereby said recirculated fluid mixture may be cooled or heated by indirect heat exchange with cooling or heating medium, respectively.

The invention will be further described by way of example with reference to the drawings accompanying the provisional specification.

Fig. 1 is a diagrammatic sectional arrangement of one embodiment of apparatus relating to the invention,

Fig. 2 is a detail of parts incorporated in the embodiment of Fig. 1,

Fig. 3 is a diagrammatic sectional arrangement of a second embodiment of apparatus relating to the invention,

Fig. 4 is a detail of parts incorporated in the embodiment of Fig. 3, and

Fig. 5 is a diagrammatic sectional arrange-

ment of a third embodiment of apparatus relating to the invention.

Referring to Figs. 1 and 2, a reaction vessel 1 is provided with an inner tubular member 2 concentric with the longitudinal axis of the vessel and open at both ends. The tubular member 2 is suitably supported inside the vessel 1 by means (not shown) and incoroporates a converging section 3 at its upper end and a diverging section 4 at its lower end. One fluid reactant under pressure is supplied through a conduit 5, another fluid reactant under pressure is supplied through a conduit 6, and the reactants mix in a conduit 7. The pressurized mixture of fluid reactants passes into an inflow connection member 8 which is connected to a tube piece 9 terminating in a nozzle 10. The inflow connection member 8 is connected by a flange 18 to a corresponding flange attached to the conduit 7, and by a flange 19 to a flange 20 attached to a top inflow connection of the vessel 1. A flange 21 attached to the tube piece 9 is clamped between the flanges 19 and 20. The nozzle 10 projects towards the upper open, converging, end of tubular member 2.

A spindle 12 is located inside the flow connection member 8 and the tube piece 9 concentrically with the axis of the nozzle 10 and is provided at its top with a guiding thread and attached handwheel 13, and at its bottom is screwed to a conically shaped nozzle constricting member 11. By manipulating the handwheel, the spindle 12 and the nozzle constricting member 11 can be moved towards or away from the throat of the nozzle 10 so that the flow cross-sectional area at the throat of the nozzle may be varied from fully closed to fully open.

An outflow conduit 14 is connected to the vessel 1 for withdrawal of product. A differential pressure meter 15 is connected to the member 8 at the point 16 and to the conduit 14 at the point 17 for measuring the pressure difference across the nozzle 10.

During the operation, the fluid reactants are supplied through the conduits 5 and 6 at the desired proportional rates of flow. The pressurized mixture of reactants passes through the nozzle 10 at a high velocity and induces recirculation of the fluid mixture through the circuit comprising the member 2 and the annular space 16 between the vessel 1 and the member 2, such fluid mixture flowing downwardly through the tubular member 2, in which reaction takes place, and upwardly through the annular space 16. The handwheel 13 of the spindle 12 is adjusted to position the nozzle closure member 11 relatively to the nozzle 10 so that the differential pressure meter 15 registers a predetermined pressure difference corresponding to a desired rate of recirculation of said fluid mixture. The reaction zone may be contained wholly within the tubular member 2 or may extend into the annular space 16.

The product of the reaction is withdrawn through the conduit 17 and a regulating valve (not shown) is provided in said conduit 17 and adjusted to maintain a predetermined pressure in the vessel 1.

When the rate of supply of fluid reactantants through, say, the conduit 5 is reduced, the rate of supply of the fluid reactant through the conduit 6 is proportionally reduced, to maintain the desired proportion of reactants, and the handwheel 13 is adjusted to maintain the predetermined pressure difference indicated on the meter 15.

Examples of reactions which can advantageously be carried out in the apparatus and according to the method described with reference to Figs. 1 and 2 are: reacting a preheated hydrogen-containing gas with a preheated gas or vapour consisting substantially of higher hydrocarbons to yield a combustible gas enriched with lower saturated hydrocarbons, reacting a hydrocarbon mixture with sulphuric acid to yield a product containing one or more sulphates or sulphonates, and effecting controlled hydrolysis by reacting various organic compounds with water or an alkali solution.

Referring to Figs. 3 and 4, a reaction vessel 31 contains a zone 32 packed with catalyst material constituting the reaction zone. The catalyst material may be supported on a grid or perforated plate (not shown). One fluid reactant under pressure is supplied through a conduit 35, another fluid reactant under pressure is supplied through a conduit 36, and the reactants mix in a conduit 37. The pressurized mixture of fluid reactants passes into an inflow connection member 38 into which is screwed a nozzle piece 40 shaped to provide the member 38 with a nozzle. The inflow connection member 38 is connected by a flange 48 to a corresponding flange attached to the conduit 37. The part of the member 38 on the downstream side of the nozzle is connected by a flange 49 to a flange 50 attached to a conduit 33 which connects with the inlet of the reaction vessel 31, and by a flange 51 to a corresponding flange attached to a recirculation conduit 34 which connects with the outlet of the vessel 31.

A spindle 42 is located inside the inlet connection member 38 concentrically with the axis of the nozzle piece 40, and is provided at one end with a guiding thread and attached to a handwheel 43, and at the other end is screwed to a conically shaped nozzle constricting member 41. By manipulating the handwheel, the spindle 42 and nozzle constricting member 41 can be moved towards or away from the throat of the nozzle so that the flow cross-sectional area

at the throat of the nozzle may be varied from fully closed to fully open.

The outlet of the vessel 31 is connected to a conduit 39 from which branch the conduit 34, through which fluid mixture is recirculated, and an outflow conduit 44, through which the reaction product is withdrawn. A differential pressure meter 45 is connected to the member 38 at point 46 upstream of the nozzle and to the conduit 33 at point 47 for measuring the pressure difference across the nozzle.

Fig. 3 shows a cooling or heating jacket 52 surrounding part of the conduit 34. A cooling or heating medium enters the jacket through a connection 53 and leaves the jacket through a connection 54.

Instead of providing a cooling or heating jacket the conduit 34 may be connected in circuit with one pass of a heat exchanger, for example of shell and tubes construction, the cooling or heating medium being connected in circuit with another pass of the heat exchanger.

During operation, the fluid reactants are supplied through the conduits 35 and 36 at the desired proportional rates of flow. The prescribed mixture of reactants passes throuh the nozzle piece 40 at a high velocity and induces recirculation of the fluid mixture through the circuit comprising the conduit 33, the reaction vessel 31 and the conduit 34. The handwheel 43 of the spindle 42 is adjusted to position the nozzle constricting member 41 relatively to the nozzle so that the differential pressure meter 45 registers a predetermined pressure difference corresponding to a desired rate of recirculation of said fluid mixture.

The reaction product is withdrawn through the conduit 44 and a regulating valve (not shown) is provided in said conduit 44 and adjusted to maintain a predetermined pressure in the vessel 31.

When the reaction is exothermic, the fluid mixture recirculated in the conduit 34 may be cooled by indirect heat exchange with a cooling medium, and when the reaction is endothermic said recirculated fluid mixture may be heated by indirect heat exchange with a heating medium.

An example of a reaction which can advantageously be carried out in the apparatus and according to the method described with reference to Figs. 3 and 4 is the strongly exothermic reaction between carbon monoxide and hydrogen to yield methane.

The reaction zone is packed with a reduced nickel catalyist supported on Kieselguhr and is maintained at a temperature within the range 250° to 270°C. by cooling the gas recirculated in the conduit 34 with a suitable cooling medium in a heat exchanger. The reactants may be contained in a single gas stream, under pressure, and supplied through say the conduit 35, the conduit 36 not being required.

Referring to Fig. 5, a reaction vessel 60 is of shell and tubes construction and comprises an upper tube plate 64 clamped between flanges of an upper shell section 62 and a main shell section 61, a lower tube plate 65 clamped between flanges of the main shell section 61 and a lower shell section 63, and a central tube 67 and a number of outer tubes 66 expanded into or rigidly attached to said tube plates. The outer tubes 66 are packed with catalyist material and constitute the reaction zone. The central tube 67 is preferably of larger diameter than the tubes 66 and serves for recirculation of fluid mixture. One fluid reactant under pressure is supplied through a conduit 68, another fluid reactant under pressure is supplied through a conduit 69 and the reactants mix in a conduit 70. The pressurized mixture of fluid reactants passes into an inflow connection member 71 which is connected to a tube piece 72 terminating in a nozzle 73. The nozzle 73 is directed towards the upper open end of the central tube 67.

A spindle 75 is located inside the inflow connection member 71 and the tube piece 72 concentrically with the axis of the nozzle 73, and is connected at its top to a diaphragm motor 76 of an automatic control system, of which a controller 78 is actuated by the pressure difference across the nozzle, and at its bottom is screwed to a conically shaped nozzle constricting member 74. By the action of the diaphragm motor 76, the spindle 75 and the nozzle constricting member 74 can be moved towards or away from the throat of the nozzle thereby decreasing or increasing the flow cross-sectional area at the throat of the nozzle.

An outflow conduit 77 is connected to the upper shell section 62 for withdrawal of reaction product. The controller 78 is provided with an indicator or recorder for the pressure difference and is connected to the member 71 at point 79 and to the conduit 77 at point 80 for measuring the pressure difference across the nozzle 73.

The main shell section 61 is connected to a conduit 81 containing a control valve 82 for inlet of a cooling or heating medium, and to a conduit 84 for outlet of the cooling or heating medium. The control valve is operated by a diaphragm motor 83 of an automatic control system, of which a controller 85 is actuated by the temperature of the fluid mixture in the upper shell section 62. The controller 85 is provided with a temperature indicator or recorder and is connected to a thermometer or pyrometer 86 located in the upper shell section 62.

During operation, the controller 78 is set to maintain a predetermined pressure difference across the nozzle, the controller

85 is set to maintain a predetermined temperature in the shell section 62 and the fluid reactants are supplied through the conduits 68 and 69 respectively at the desired proportional rates of flow. The pressurized mixture of reactants passes through the nozzle 73 at a high velocity and induces recirculation of the fluid mixture through the circuit comprising the central tube 67, the lower shell section 63, the reaction zone tubes 66 and the upper shell section 62 automatically at a desired rate corresponding to the set predetermined pressure difference, such fluid mixture flowing downwardly through the central tube 67 and upwardly through the reaction zone tubes 66. The flow of cooling or heating medium through the space surrounding the tubes in the main shell section 61 is automatically controlled by the valve 82 to maintain the set predetermined temperature measured by the thermometer or pyrometer 86. A cooling medium is used when the reaction is exothermic and a heating medium when the reaction is endothermic.

An example of a reaction which can advantageously be carried out in the apparatus and according to the method described with reference to Fig. 5 is the strongly exothermic reaction when naphthalene is oxidised by air to yield phthalic anhydride. The tubes 66 are packed with a vanadium pentoxide catalyst supported on an inert refractory material and the temperature measured by the pyrometer 86 is maintained within the range 350° to 450°C., using a suitable cooling medium supplied through the conduit 81. A mixture of naphthalene vapour and preheated primary air is supplied through the conduit 68 and secondary air is supplied through the conduit 69.

WHAT WE CLAIM IS :—

1. A method of continuously reacting fluids with one another comprising injecting a pressurized mixture of fluid reactants through at least one nozzle, having a throat with a variable flow cross-sectional area, and into a circuit, comprising a reaction zone connected by its outlet back to its inlet, to induce recirculation of the fluid mixture through said reaction zone and around said circuit, adjusting the flow cross-sectional area of said nozzle throat to obtain a desired rate of recirculation of said fluid mixture, and withdrawing a proportion of said recirculated fluid mixture as reaction product.

2. A method as claimed in claim 1 wherein the flow cross-sectional area of said nozzle is adjusted automatically in response to the pressure drop across the nozzle.

3. A method as claimed in claim 1 or 2 wherein the reactants are supplied as separate streams which are mixed at the desired proportional rates of flow near the vicinity of the nozzle.

4. A method as claimed in claim 3 wherein at least one of the reactant streams is preheated.

5. A method as claimed in any preceding claim wherein the reaction zone contains a catalyist material for promoting the reaction.

6. A method as claimed in any of claims 1 to 5 wherein the reaction is exothermic and the fluid mixture being recirculated is cooled by indirect heat exchange with a cooling medium.

7. A method as claimed in any of claims 1 to 5 wherein the reaction is endothermic and the fluid mixture being recirculated is heated by indirect heat exchange with the heating medium.

8. A method of continuously reacting fluids with one another substantially as herein described with reference to Figs. 1 and 2 of the drawings accompanying the provisional specification.

9. A method of continuously reacting fluids with one another substantially as herein described with reference to Figs. 3 and 4 of the drawings accompanying the provisional specification.

10. A method of continuously reacting fluids with one another substantially as herein described with reference to Fig. 5 of the drawings accompanying the provisional specification.

11. Apparatus for continuously reacting fluids with one another comprising a reaction vessel which contains a reaction zone connected by its outlet back to its inlet to form a circuit, at least one fluid-reactant inflow connection member terminating in an injector nozzle arranged in said circuit for the injection of a mixture of fluid reactants into said reaction zone in such a manner as to induce the recirculation of fluid mixture around said circuit and through said reaction zone, means for varying the flow cross-sectional area at the throat of said nozzle and an outflow connection member from said circuit for reaction product.

12. Apparatus as claimed in claim 11 wherein said means for varying the throat area of the nozzle comprises a suitably shaped nozzle constricting member attached to a spindle concentric with the axis of the nozzle, said spindle being movable along said axis towards and away from the throat of the nozzle, so that the flow cross-sectional area at the throat of the nozzle may be varied from fully closed to fully open.

13. Apparatus as claimed in claim 12 wherein said spindle is screw-threaded and is attached to a hand wheel for manual adjustment of the nozzle constricting member.

14. Apparatus as claimed in claim 12 wherein said spindle is connected to a motor of an automatic control system for automatic adjustment of the nozzle constricting

member in response to the pressure drop across the nozzle.

15. Apparatus as claimed in any of claims 11 to 14 wherein said nozzle is rigidly and detachably attached to said inflow connection member.

16. Apparatus as claimed in any of claims 11 to 15 wherein the reaction vessel is partitioned so that the circuit for recirculation is wholly within the reaction vessel.

17. Apparatus as claimed in claim 16 wherein the reaction vessel has an inner tubular member concentric with a longitudinal axis of the vessel and open at both ends, said nozzle being directed concentrically towards an inlet end of said tubular member.

18. Apparatus as claimed in claim 17 wherein said inner tubular member incorporates a convergent section at its inlet end and a divergent section at its other open end forming its outlet.

19. Apparatus as claimed in claim 16 wherein the reaction vessel comprises a shell with tube plates rigidly attached thereto at opposite ends and a number of open-ended tubes expanded into or otherwise rigidly attached to said tube plates and extending from one tube plate to the other, a central tube serving for recirculation of fluid mixture and outer tubes constituting the reaction zone, said nozzle being inserted into said reaction vessel to be directed towards said central tube and concentric therewith, the reaction vessel having inlet and outlet connections for a cooling or heating medium for passage through the space outside said tubes and between the tube plates.

20. Apparatus as claimed in claim 19 wherein said outer tubes contain a catalyst material.

21. Apparatus as claimed in any of claims 11 to 15 wherein the whole or a major part of the reaction vessel constitutes the reaction zone and wherein a recirculation conduit branches from said outflow connection member and leads back to an inlet to the reaction vessel, said nozzle being directed into said recirculation condiut, preferably close to the inlet to the reaction vessel, for the recirculation of fluid mixture.

22. Apparatus as claimed in claim 21 wherein the reaction vessel contains a catalyst material.

23. Apparatus as claimed in claim 21 or 22 wherein an indirect heat exchanger is incoropated in said recirculation conduit.

24. Apparatus for continuously reacting fluids with one another constructed and adapted to operate substantially as herein described with reference to and as illustrated in Figs. 1 and 2 of the drawings accompanying the provisional specification.

25. Apparatus for continuously reacting fluids with one another constructed and dapted to operate substantially as herein described with reference to and as illustrated in Figs. 3 and 4 of the drawings accompanying the provisional specification.

26. Apparatus for continuously reacting fluids with one another constructed and adapted to operate substantially as herein described with reference to and as illustrated in Fig. 5 of the drawings accompanying the provisional specification.

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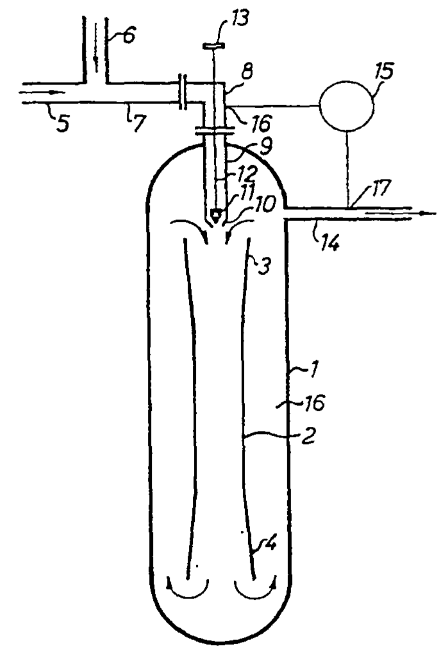

FIG. 1.

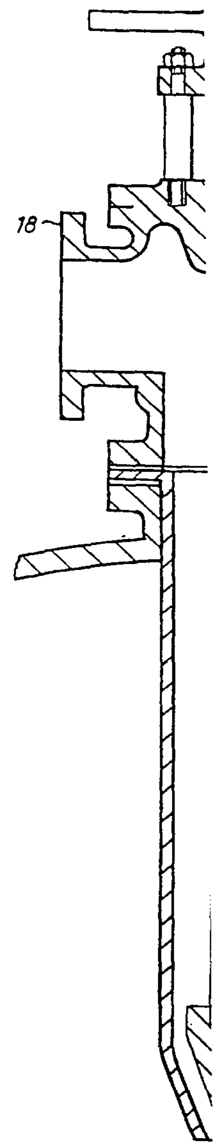

FIG.

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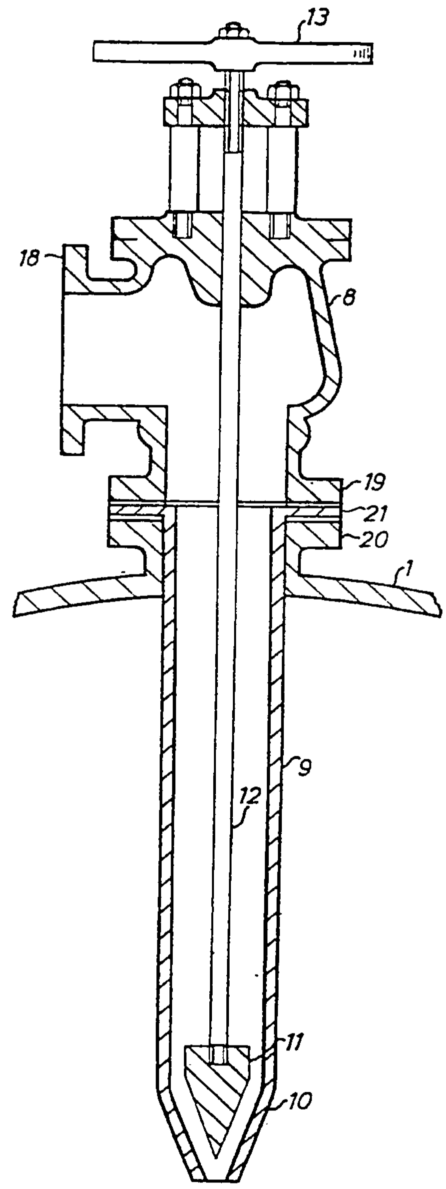

FIG. 2.

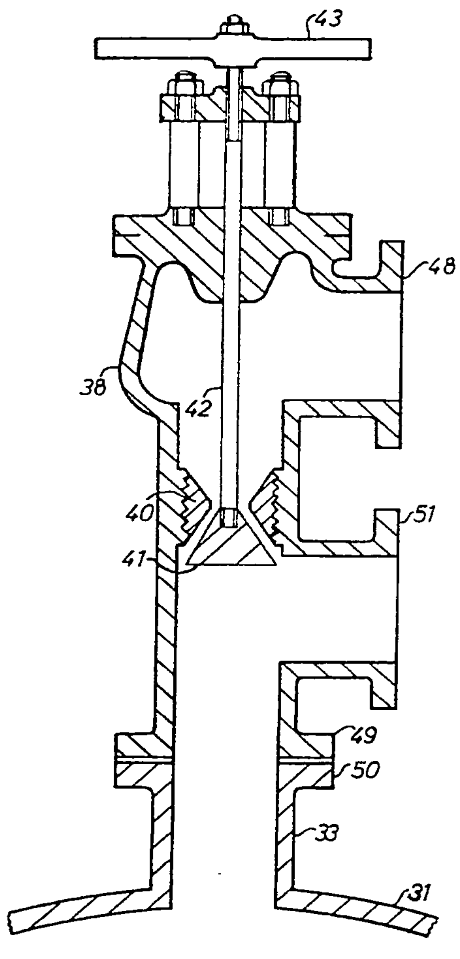

FIG. 4.

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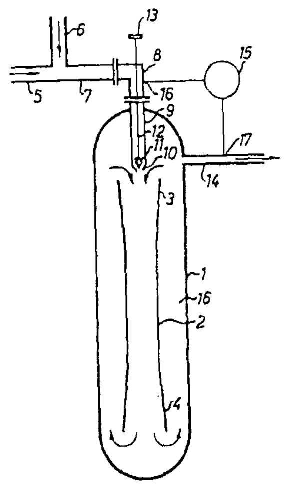

FIG. 1.

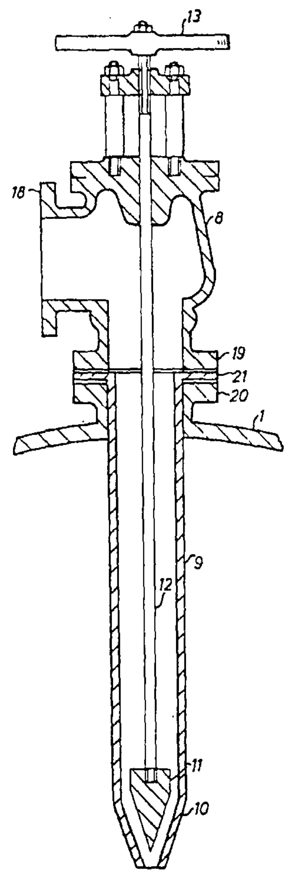

FIG. 2.

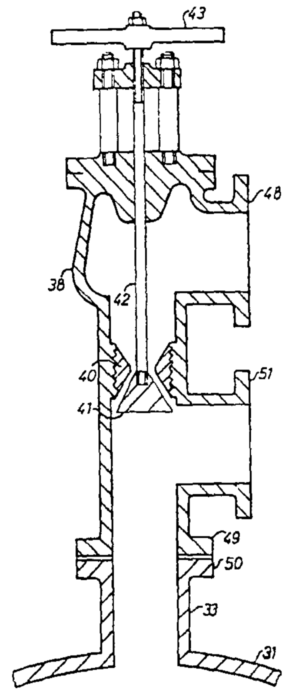

FIG. 4

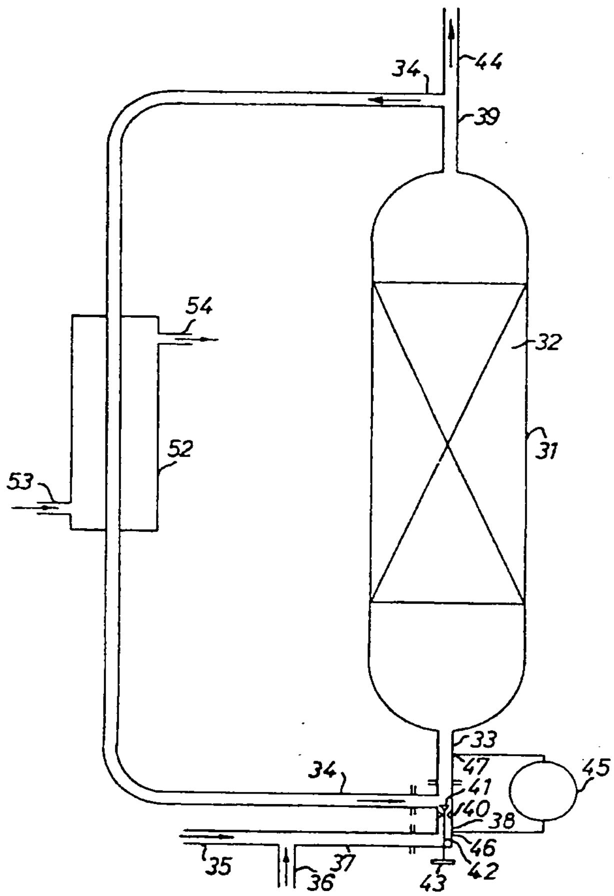

FIG. 3.

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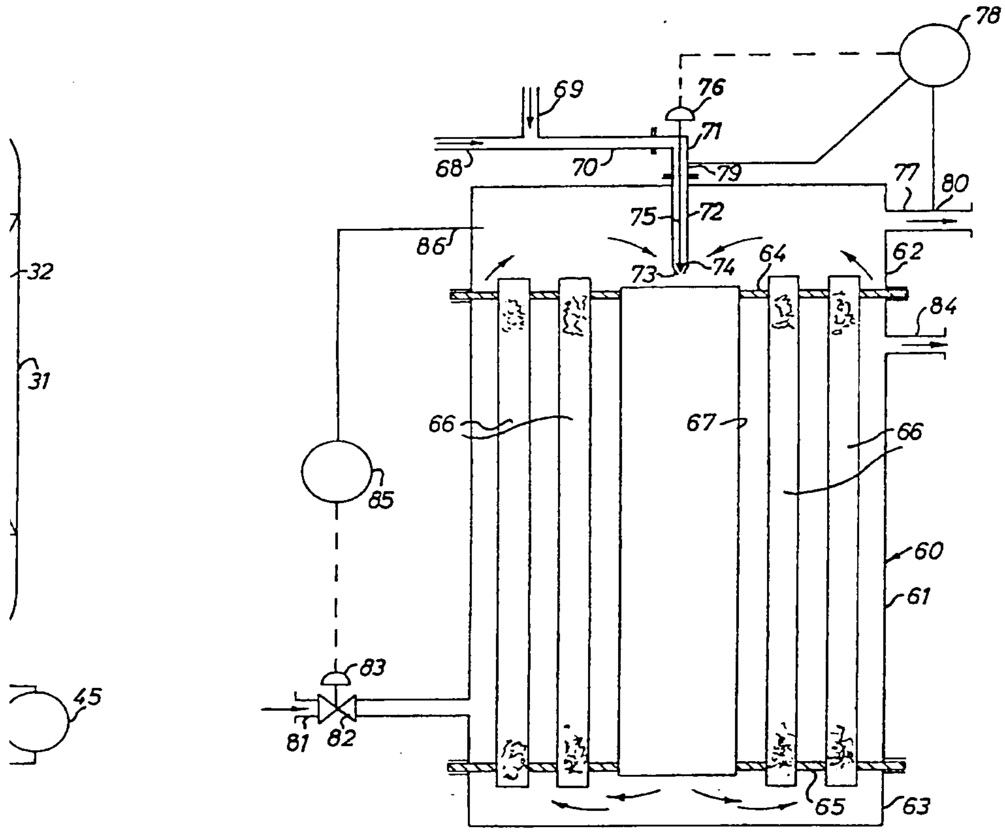

FIG.5.

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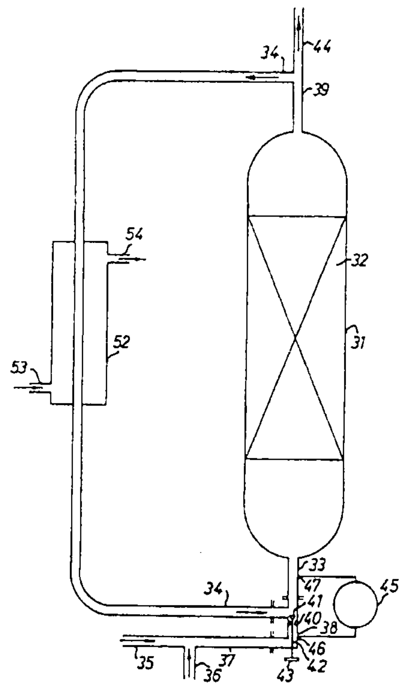

FIG. 3

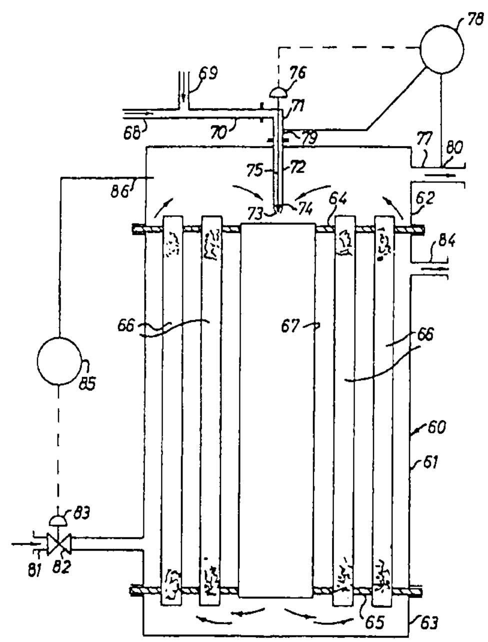

FIG. 5.